NUMERICAL CALCULATIONS OF STRATOSPHERIC OZONE TRANSPORT

by

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THESIS

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ABSTRACT

The vertical distribution of ozone was observed by the Air Force Cambridge Research Laboratories North American ozonesonde network from 29 April 1963 to 10 May 1963 on a daily basis. This observed data, supplemented by twenty-four hour trajectory calculations, was used to prepare distributions of ozone mixing ratio on the 200, 100, 50, and 30 mb surfaces. The ozone distributions were evaluated for the ozone transport by mean and eddy motions in the lower stratosphere. The results of these calculations show the magnitude of the horizontal and vertical transport of ozone into and out of the 30/50, 50/100, and 100/200 mb volume over North America. The mean vertical transport, supported by the vertical eddy transport, is combined to move the ozone primarily downward through the stratosphere. At the tropopause the vertical eddies transport the ozone to the troposphere for eventual destruction.

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LIST OF SYMBOLS

A	= area enclosed within lateral boundary of computational grid
c_p	= specific heat of air at constant pressure
H	= heat per unit mass
i	= grid index in east-west direction, positive eastward
j	= grid index in the north-south direction, positive northward
K_H	= $K_x = K_y$
K_p	= eddy-diffusion coefficient of χ in p direction
K_x	= eddy-diffusion coefficient of χ in x direction
K_y	= eddy-diffusion coefficient of χ in y direction
l	= distance around the lateral boundary of computational grid
n	= coordinate direction perpendicular to lateral boundary of computational grid, defined positive inward
p	= air pressure in millibars
Q	= a dummy variable used for illustrative purposes
$\overline{Q}$	= average along a horizontal line
$\tilde{Q}$	= area average $\equiv \frac{1}{A}\iint Q \, dA$
$\tilde{\tilde{Q}}$	= vertical average $\equiv \frac{1}{\Delta p}\int Q \, \delta p$
T	= temperature in degrees absolute
t	= time
u	= zonal component of the wind
$\mathbb{V}_2$	= wind velocity
v	= meridional component of the wind
$\overline{(v'w')}$	= covariance of the meridional and vertical eddy components of the wind along a latitude circle

w = vertical motion in height coordinates = $\frac{dz}{dt}$

α = specific volume

χ = mixing ratio of ozone expressed as micrograms per gram

ω = vertical velocity of wind in pressure coordinates = $\frac{dp}{dt}$

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I. INTRODUCTION

Ozone is an important but not well-documented atmospheric gas. Its importance to meteorologists is twofold. First, ozone can be used as a quasi-conservative atmospheric trace substance below 30 kilometers and, second, as an absorber of ultraviolet light, ozone affects the heat balance through the diabatic heating term of the first law of thermodynamics. Although the exchange of energy by ozone is worthy of investigation, ozone as an atmospheric tracer is the subject of this study. Before proceeding, a review of the significant works of earlier investigators is desirable.

The first major investigation of the measurement and distribution of atmospheric ozone was carried out under the direction of G. M. B. Dobson during a five-year period from 1924 to 1929. Dobson and his associates reported the findings of their study in a series of four articles in the Proceedings of the Royal Society of London (1926, 1927, 1929, 1930). Based on the work of previous investigators, Dobson developed a spectroscopic instrument for use on the earth's surface that measures the total amount of atmospheric ozone in a vertical column. The results of daily measurements of total ozone at Oxford during 1924 and 1925 were compared with surface pressure. It was found that surface pressure had a high negative correlation, on the order of -.5, with total ozone measurements. On an annual basis, a maximum of total ozone was observed in the spring with a corresponding minimum in the fall. Comparisons of total ozone were made with atmospheric pressure at nine, twelve, and fourteen kilometers. A high negative correlation, on the order of -.8, was observed.

In 1926 instruments were sent to various locations in northwestern Europe. The observations produced similar conclusions as the Oxford findings of 1925. Using the results of 1926-1927 a complete description of the ozone conditions relative to cyclones and anticyclones was presented. In general, high total ozone was measured in the area of northerly wind flow and low total ozone in the sectors associated with southerly wind flow. Highest total ozone values were found to the southwest of the surface low pressure centers.

During 1928 and 1929 the spectrographs were redistributed so that observations were taken at Arosa (Switzerland), Table Mountain (California), Helwan (Egypt), Kodaikanal (India), Christchurch (New Zealand), Oxford (England), and Montezuma (Chile). The analysis of the measurements showed that the midlatitude stations had similar observations as discussed for northwestern Europe. The tropical stations observed no significant pressure or total ozone variations.

Several years later, using the observations obtained during the period 1928-1932 in northwestern Europe, Meetham (1937) found a high positive correlation between total ozone and temperature at a height of 12, 15, and 18 kilometers. The magnitude of the correlation coefficients was on the order of +.5. He also found a high negative correlation between total ozone and pressure at 9, 12, 15, and 18 kilometers altitude and between total ozone and the height of the tropopause. Although values vary somewhat, the pressure correlation coefficients have a magnitude of -.4, while the corresponding tropopause figures are -.5.

Godson (1960) has summarized Northern Hemisphere total ozone observations over many years by month and latitude. His results show a strong middle and north latitude spring maximum during the month of March and a middle and north latitude minimum during the month of October. Relatively constant values over the seasons are shown in the tropics with increasing values northward. The maximum value is reported above 70N during the month of March. The steepest latitudinal gradient occurs across middle latitudes during late winter and early spring. The flattest gradient occurs during late summer and early autumn. This elaborates upon and confirms earlier and limited observations. Godson's summary is generally accepted as a reasonable description of the mean distribution of total ozone.

The vertical distribution of ozone has been difficult to obtain. This is attributed to the lack of a reliably accurate, inexpensive, and expendable instrument. A summation of the various methods of measuring the vertical distribution of ozone is presented by Craig (1965) and therefore is not presented here. The most widely used technique has been the <u>Umkehr</u> method devised by F. P. W. Gotz in 1931 (Craig, 1965). Using photometery, this method breaks the atmosphere into five or nine layers. For each layer a single ozone value is computed. Of more recent use are the Regener chemiluminescent ozonesonde and the Brewer-Mast ozonesonde. Both of these instruments are capable of being used on radiosondes and are able to give a much finer vertical ozone structure than the <u>Umkehr</u> method.

Investigators (see, for example, Gotz, 1951; Ramanathan and Kulkarni, 1960; Kulkarni, 1962; Mateer and Godson, 1960; Craig, 1960, 1965; Hering and Borden, 1965; or Bojkov, 1965, 1969) agree on the general shape of the vertical ozone profile. Low, relatively constant, mixing ratios of ozone are found in the troposphere with a very sharp increase at the tropopause. The maximum value of ozone mixing ratio is most often found in the 15 to 30 kilometer layer with the altitude of the maximum decreasing poleward. Above this maximum the ozone mixing ratio slowly decreases or remains relatively constant with altitude. Maximum hemispheric values are observed near the 30-kilometer altitude of the equatorial latitudes. In the stratosphere isolines of ozone distribution normally slope poleward and downward from the tropical regions. Large daily fluctuations in ozone content are observed in the region between the tropopause and the primary ozone maximum. The fluctuations are weakest in tropical regions and strongest in middle and north latitudes.

It is worthwhile at this point to consider the expected distribution of ozone assuming photochemical equilibrium. Gotz (1951), Craig (1950, 1965), Prabhakara (1963), and Brewer and Wilson (1968) have discussed the origin and details of photochemical theory with respect to atmospheric ozone. Therefore, photochemical theory will not be reviewed here. From the work of the previously noted authors, two salient points are noted:

1. Photochemical equilibrium theory correctly predicts the observed amount of ozone in the atmosphere above approximately 30 kilometers. Predicted by this theory is the equatorial maximum region and decreasing values poleward and with altitude. Below this region, photochemical theory does not agree with observed distributions.

2. Production and recombination can be considered small below the 30 kilometer maximum observed values. Therefore in this region

ozone must be quasi-conservative and can be used as an atmospheric tracer.

Since the observed distribution and photochemical theory disagree, the discrepancy must be due to transport processes. Transport mechanisms must move the ozone from high equatorial source regions to the lower stratosphere in middle and high latitudes. From the lower stratosphere ozone is passed to the troposphere for eventual destruction with atmospheric constituents or the earth's surface (Craig, 1950, 1965). Craig further mentions that the process would have to be "most effective in middle and high latitudes in winter, when the total amount of ozone increases most rapidly" (Craig, 1950).

Prabhakara (1963) performed a study of a mathematical model of ozone distribution based on photochemical equilibrium below 41 kilometers and the effect of transport processes upon this distribution. His conclusion that "photochemical theory by itself cannot explain the latitudinal and seasonal variation of ozone" lends further support to ideas advanced by Craig (1950, 1965).

In the past, sufficient ozone measurements on a hemispheric and vertical scale were unavailable. This prevented direct calculations of the transport of ozone by dynamic processes in the stratosphere. Newell (1961, 1962, 1964) advanced the idea that since the total ozone measured at the surface was highly correlated with the ozone amount in the 12 to 24 kilometer layer, then a measurement of the flux of total ozone would be indicative of the flux in the lower stratosphere. Using 100 and 50 mb winds and total ozone measurements during the International Geophysical Year, Newell computed the mean and eddy fluxes of ozone in this layer. Newell concluded that the mean meridional circulation and standing eddies did not serve as effective transport mechanisms. Thus, he argued that most ozone transport was attributed to quasi-horizontal transient eddies.

To explain the observed change in total ozone measured at the surface, a mechanism was proposed by Reed (1950) and Normand (1953). Although proposed separately, this theory is usually called the "Reed-Normand effect". The mechanism combines on a synoptic scale the horizontal advection and vertical motion to explain the variation of total ozone with the synoptic features of the upper troposphere and lower stratosphere. Basically the argument is as follows.

Air blows through the ridge and trough patterns of the height field. The temperature pattern associated with the height field in the lower stratosphere is fairly constant with warm areas in the base of troughs and cold areas in the ridges. Therefore, the quasi-stationary temperature patterns must be maintained by adiabatic warming and cooling of air parcels moving through them. The subsidence associated with the warming brings down ozone-enriched air from above. The lifting, associated with the cooling, transports ozone-poor air from lower altitudes. Since this region is below the level of the observed equatorial maximum, higher ozone values are brought southward to the trough and lower values are brought northward from equatorial regions to the ridge.

The basic qualitative theories of movement and approximate distribution of ozone in the lower stratosphere have been proposed. Quantitative conformation of these theories has been lacking due to a minimum of information with respect to the horizontal and vertical distribution of ozone in the lower stratosphere (and elsewhere). With the challenge thus defined, this study was undertaken to compare earlier theories with a quantitative description of ozone movements.

As a first step the distribution of ozone mixing ratio on standard pressure surfaces in the lower stratosphere was constructed from the observations taken by the Air Force Cambridge Research Laboratories ozonesonde network during 29 April 1963 to 10 May 1963. It follows that ozone motions can be evaluated using basic meteorological data.

To accomplish this evaluation, a computational scheme was proposed by Professor J. D. Mahlman of the Department of Meteorology, Naval Postgraduate School. This scheme puts the continuity equation for ozone (Haltiner and Martin, 1957) into a form that enables computation of the mean and eddy flux of ozone mixing ratio into a volume bounded by two pressure surfaces. The intent of the study is to perform quantitative calculations of ozone transport due to various physical processes based upon this formulation and assess their significance in terms of previous estimates.

II. PREPARATION OF DATA AND THE COMPUTATIONAL SCHEME

A. BACKGROUND

In 1963 an attempt was made to gather a more complete representation of the distribution of ozone on a synoptic scale. The Air Force Cambridge Research Laboratories (AFCRL) operated an eleven-station ozonesonde network in North America. Daily observations obtained by this network between 29 April 1963 and 10 May 1963 provide the basic data for this study (Hering and Borden, 1964). This period is just after the observed spring maximum of total ozone. Wind and temperature fields at standard levels were available from the National Weather Records Center, Asheville, North Carolina and from the Free University of Berlin (1963, 1966).

B. PHYSICAL LIMITATIONS

Considering the geographical location of the ozonesonde stations on the North American continent, an upper boundary of 24 kilometers defined by sufficient ozonesonde observations, and a lower boundary defined by the tropopause, the volume in which calculations can be made is limited.

Four pressure levels - 200, 100, 50 and 30 mb - were chosen on the basis of readily available wind and temperature data. The selection of the grid boundaries at 30N-140W, 75N-140W, 75N-60W, and 30N-60W minimizes the Atlantic Ocean, Pacific Ocean, and Northern Canada data-sparse areas.

A latitude-longitude oriented grid was used in order to measure fluxes across boundaries of meridians and parallels. The grid interval on a pressure surface is five degrees, which is sufficient to represent synoptic scale motions in the stratosphere.

For the four isobaric surfaces involved the distance between them is on the order of four kilometers. Considering the vertical distribution of ozone this interval is larger than desired and forces rough approximations in computing vertical derivatives.

The time interval was chosen as one day corresponding to the frequency of ozonesonde observations. Considering the expected movement of ozone in this region and the synoptic scale of the analysis, this interval is reasonable.

C. BASIC ATMOSPHERIC PARAMETERS

Hand analyses of streamline, isotach, and isotherm patterns were prepared at each pressure surface for the time period of the calculations. The daily 200 and 100 mb 1200 GMT Weather Bureau synoptic analyses and the daily 50 and 30 mb 0000 GMT synoptic analyses from the Free University of Berlin were used as a plotted data source.

As shown in Appendix A, the streamline analyses describe typical patterns for this time of year. The lower levels reflect deep tropospheric systems that weaken with altitude. At the 30 mb surface the weakening winter circumpolar vortex is observed with weak anticyclonic centers forming and dissipating at the southern latitudes. The ten-day period of this investigation's major calculation is during the spring reversal of the upper stratospheric circulation from winter westerlies to summer easterlies.

Since the ozonesonde observations were taken at 1200 GMT, an approximation to the parameters of the atmosphere at the 50 and 30 mb surfaces was made as follows. The zonal and meridional components of the wind field and the temperature field were linearly interpolated for 1200 GMT values from the 0000 GMT fields.

Vertical motions were computed by solving the first law of thermodynamics in the form

$$\omega = \frac{\frac{\partial T}{\partial t} + \mathbb{V}_2 \cdot \nabla T - \frac{1}{c_p}\frac{dH}{dt}}{\frac{\alpha}{c_p} - \frac{\partial T}{\partial p}}. \tag{1}$$

The diabatic heating term was evaluated using the net heating rates as proposed by Kennedy (1964) for the spring period. Unrepresentative changes in vertical motions between grid points and excessive magnitudes at certain grid points are the result of computational inaccuracies attributed to the assumption of a linear change of wind and temperature across a horizontal grid interval. To avoid these unrepresentative and dynamically untenable vertical motions, a smoothing scheme was used as follows:

$$\omega_{i,j} = .15\omega_{i,j+1} + .15\omega_{i,j-1} + .15\omega_{i-1,j} + .15\omega_{i+1,j} + .4\omega_{i,j}. \tag{2}$$

To aid in establishing the state of the atmosphere for the area of interest during the period of the calculations, ten day latitudinal averages, by pressure surface, were prepared for the temperature field and the zonal, meridional, and vertical components of the wind fields. These values are presented in meridional cross section format as figures 1 - 4.

The values in figures 1 - 4 are in general agreement with results presented by other authors (e.g. Craig, 1965). The temperature field (fig. 1) is typical for the lower stratosphere at this time of year. Cold temperatures at southern latitudes give way to the warm

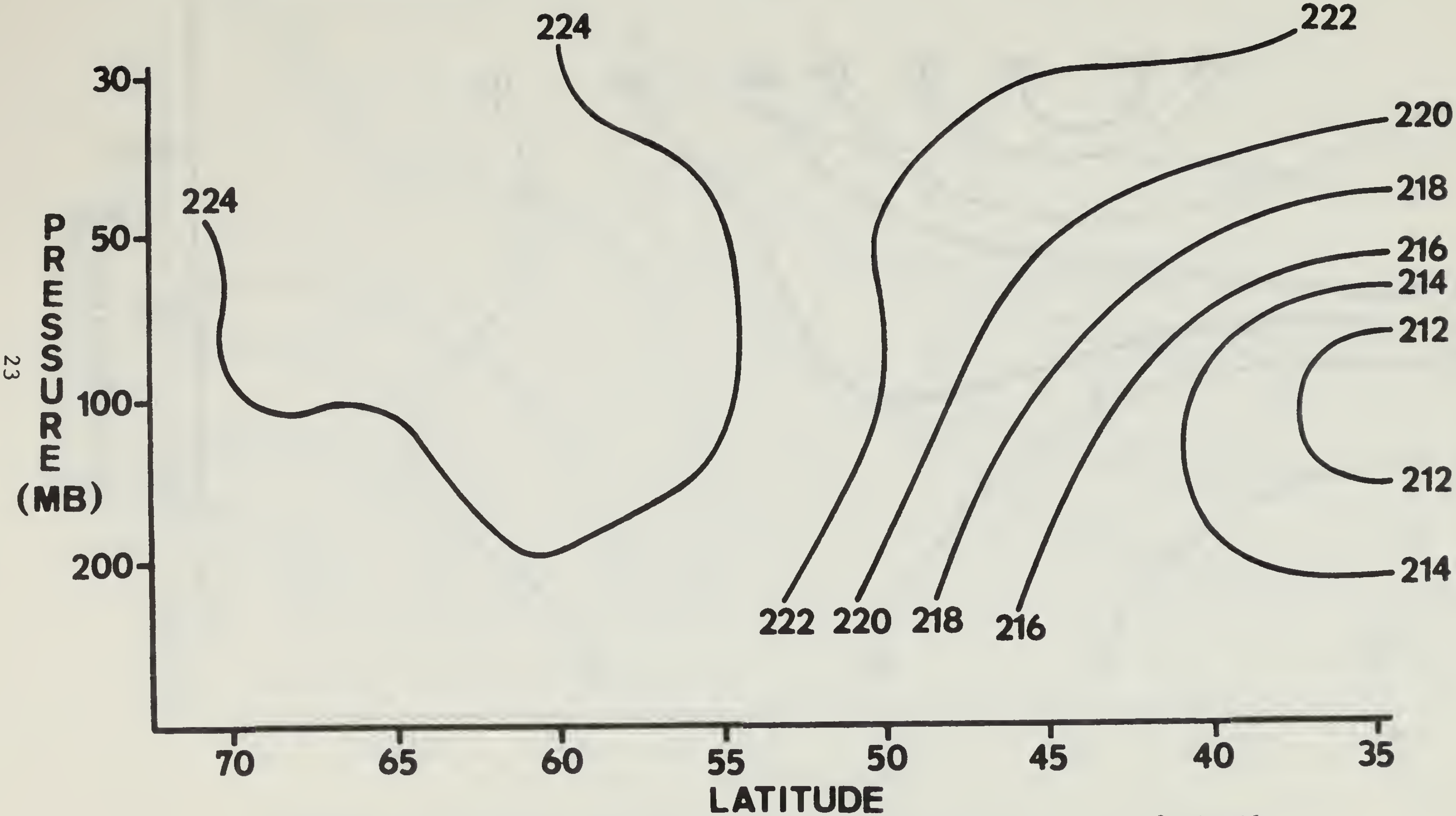

FIG. 1. Ten-day Zonal Mean Temperatures (deg K) for the period 30 April to 9 May 1963. Computed between longitude 60W and 140W.

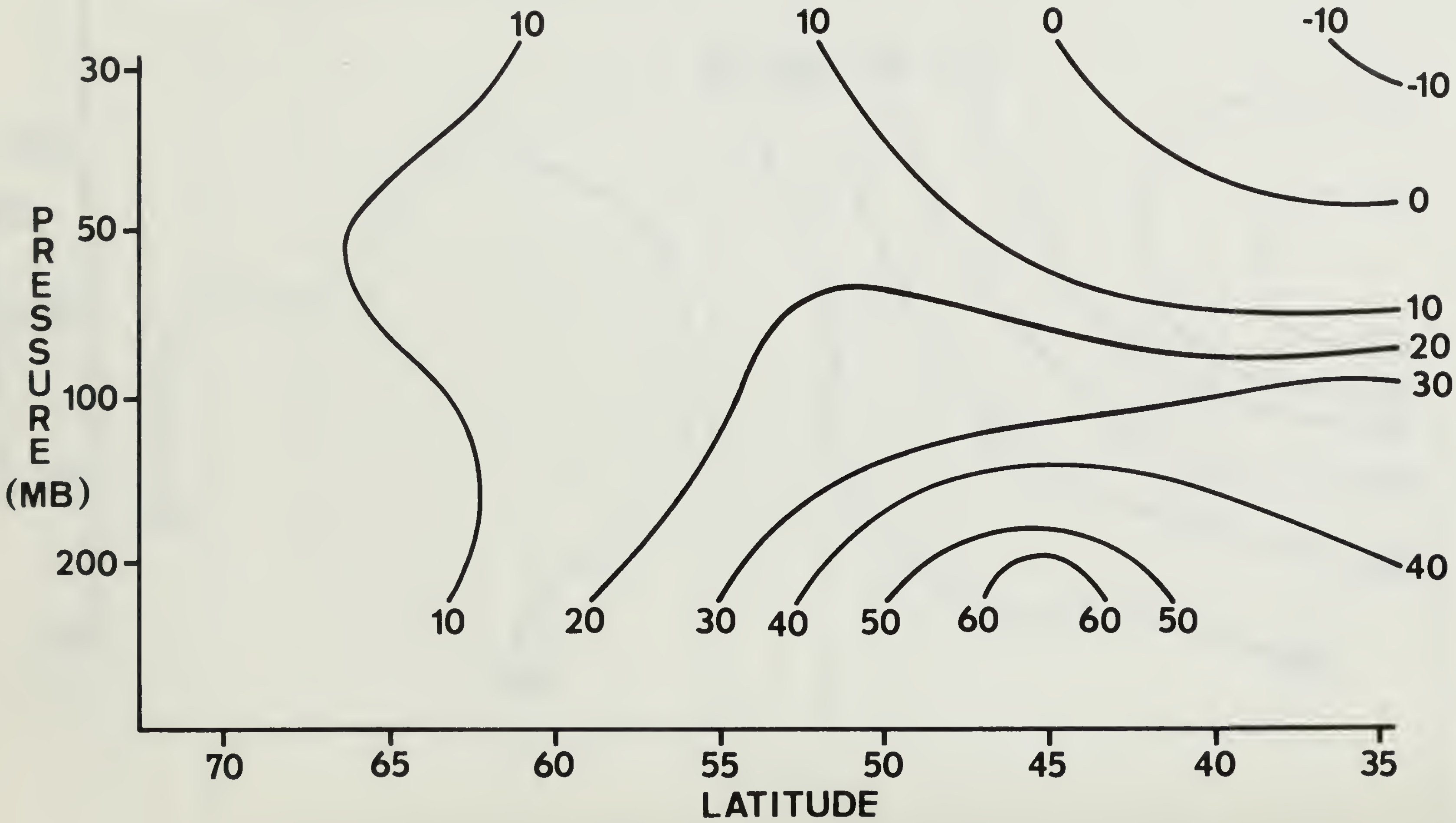

FIG. 2. Ten-day Zonal Mean u-component of Wind (Knots) for the Period 30 April to 9 May 1963. Computed between longitude 60W and 140W.

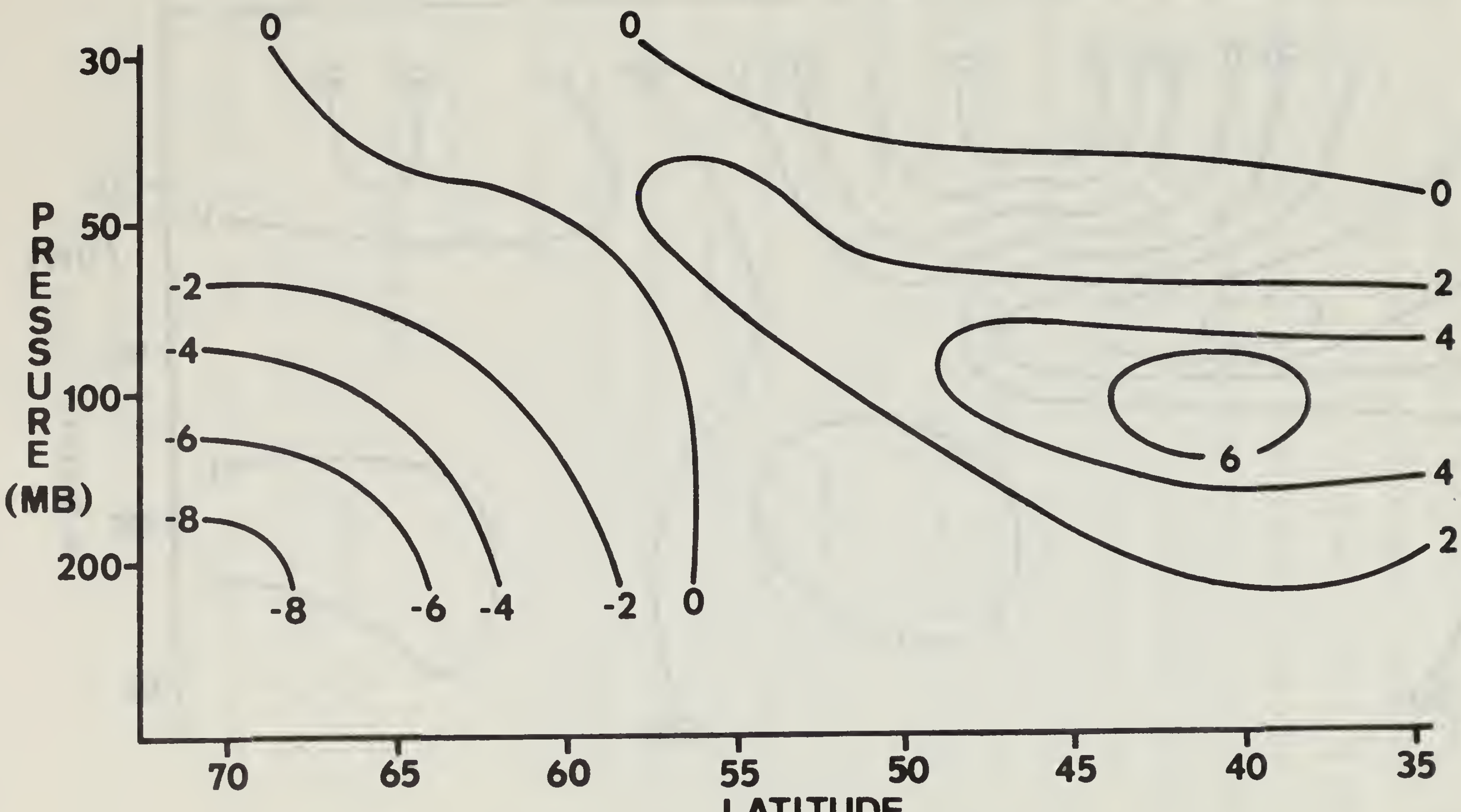

FIG. 3. Ten-day Zonal Mean v-component of Wind (Knots) for the Period 30 April to 9 May 1963. Computed between longitudes 60W and 140W.

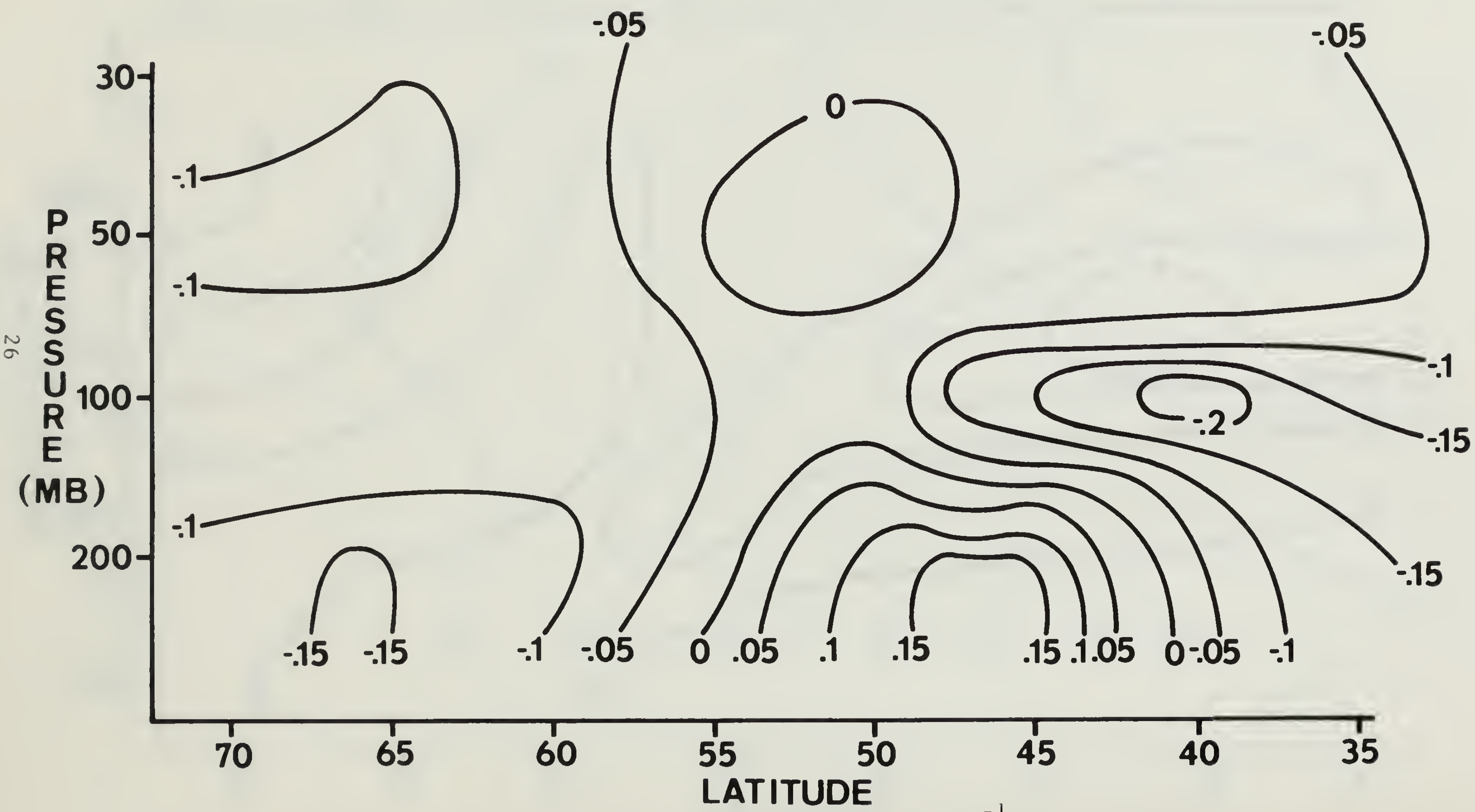

FIG. 4. Ten-day Zonal Mean Vertical Motion (km day^{-1}) for the period 30 April to 9 May 1963. Computed between longitudes 60W and 140W. Positive values indicate upward motion.

temperatures associated with the middle and northern latitude troughs. This latitudinal temperature gradient weakens with altitude. The u-component of the wind (fig. 2) supports the earlier observation that the winter circumpolar vortex is weakening and being replaced from above by summer easterlies as can be seen at 30 mb and 35N. The mid-latitude wind maximum associated with the mean polar front jet position is seen at 200 mb and 45N. The v-component of the wind (fig. 3) shows little motion at the 30 and 50 mb levels. At 100 mb strong southerly movement north of 65N and strong northerly movement south of 50N is observed. At 200 mb, strong southerly motions are seen north of 65N. The vertical motions (fig. 4) show a general subsidence over the volume of calculations except for a weak rising area at 50 mb and strong upward motions between 45N and 50N at 200 mb.

Most investigators presenting similar results use time scales of months or seasons. This study is limited to ten days and hence the values are not directly comparable. In addition, to properly discuss the various details of these mean atmospheric parameters, a separate and more thorough treatment is required than can be presented here. Therefore, further discussion of figures 1 - 4 is not presented.

D. OZONE DATA AND ANALYSIS

1. The AFCRL Ozonesonde Network

For most of its duration the AFCRL ozonesonde network made weekly observations on Wednesday except for selected periods when more frequent observations were made. During one such period from 29 April 1963 to 10 May 1963, daily observations were taken at 1200 GMT. Participating stations are listed in Table I.

OZONESONDE NETWORK

(after Hering and Borden, 1964)

STATION	LATITUDE (Deg. N)	LONGITUDE (Deg. W)
Albrook Field, Canal Zone (AWS)	9.0	79.6
Colorado State University, Fort Collins	40.6	105.1
Eielson AFB, Fairbanks, Alaska (AWS)	64.8	147.9
Florida State University, Tallahassee	30.4	84.3
Fort Churchill, Manitoba (Canadian Met. Br.)	58.8	94.1
Goose Bay, Labrador (Canadian Met. Br.)	53.3	60.4
L. G. Hanscom Field, Bedford, Massachusetts	42.5	71.3
Thule AFB, Greenland (AWS)	76.5	68.8
University of New Mexico, Albuquerque	35.0	106.6
University of Washington, Seattle	47.4	122.3
University of Wisconsin, Madison	43.1	89.4

TABLE I

The instrument used in determining the amount of ozone in the atmosphere was the dry chemiluminescent ozonesonde developed by Regener (1960). The ozonesonde was attached to a sounding balloon with a standard radiosonde package. The instrument measures the amount of ozone by drawing ozone-laden air across a disk coated with a chemical that produces a luminescent reaction with ozone. The intensity of the reaction is measured by a photomultiplier tube and in turn is telemetered back to ground using standard radiosonde equipment (Regener, 1960, 1964). Samples were taken every 15 seconds for a vertical resolution of approximately 250 feet (Hering and Borden, 1964).

2. Reliability of Measured Chemiluminescent Ozonesonde Data

In view of the uncertainties inherent with the measurement and calibration of the Regener ozonesonde it is worthwhile to note the major corrections applied to the soundings and to note the results of several intercomparison tests with other ozonesondes.

The disk coated with the chemiluminescent substance is sensitive to light, moisture, and ozone. Therefore, as a check, calibration shortly before launch is required (Regener, 1964). Hering and Borden (1964) report that achievement of reliable preflight calibration was difficult. This in turn required a correction to the telemetered data. The correction was achieved by adjusting the integrated observed ozone density to a simultaneously observed total ozone measurement using a spectrophotometer. The ozonagrams published by Hering and Borden (1964) have this correction applied.

Hering and Dütsch (1965) compared the results of simultaneous soundings of the Regener chemiluminescent ozonesonde and the bubbler-type electrochemical ozonesonde. Their conclusion is that "ozonesondes of the types tested are capable of providing reliable high-resolution measurements of the vertical ozone distribution".

More recently Komhyr, Grass, and Proulx (1968) made intercomparison tests in various combinations between the Regener chemiluminescent ozonesondes, carbon-iodine ozonesondes, and Brewer-Mast ozonesondes. The result of the comparisons was that, after corrections, the chemiluminescent ozonesonde compared with the other ozonesondes in the stratosphere and gave "relatively reliable" results.

Hering and Borden (1964) considered the data obtained by the ozonesonde network as "provisional". Considering the results of the two intercomparison tests and the observation that other investigators (Berggren and Labitzke, 1966, 1968; Breiland, 1964, 1965, 1967, 1968; Craig, DeLuisi, and Sticksel, 1967; Hering, 1966; Hering, Touart, and Borden, 1967; and Sticksel, 1966) have published studies using the corrected ozonesonde data, the reliability of the ozonesonde soundings as published by Hering and Borden (1964) is accepted.

3. Preparation of Ozone Data for Analysis

Although the ozone soundings were accepted there are several features of the soundings that had to be modified to be consistent with the scale of the intended calculations. On the ozonagram thin layers with large unrepresentative gradients and layers in which the ozone values fluctuated excessively and with apparent randomness were considered not representative of the synoptic scale distribution. These features of the vertical distribution may either be a failure of the instrument or a smaller-scale ozone distribution which is presently below our ability to identify. To overcome these apparent inconsistencies a subjective smoothing of the individual soundings was completed as the first step in analysis. This smoothing is not to be over-emphasized as extreme care was taken not to destroy the characteristics of the soundings. The severest smoothing was on the order of .3 kilometers on the height scale with 95 percent of the ozonagram values being unaffected by the smoothing process.

With the smoothed values obtained, a time series for the twelve-day period was prepared for each station. Linearly interpolated values were calculated for stations missing an observation but having an observation 24 hours prior to and after the missing observation. Linear interpolations were also made in the case of the Fairbanks station where observations were reported at 0000 GMT.

To supplement the small number of observations for the area under consideration, hand trajectories were calculated for the 24-hour periods prior to and following an observation. This technique used 12-hour streamline and isotach patterns to obtain the position of the parcel 24 hours before and after the observation time. Under conservation of ozone mixing ratio (χ), these new points then possess a value of χ corresponding to the value measured at the original point. An estimate of the vertical motion during the trajectory was made using an approximation derived from the Eulerian expansion of the total derivative of pressure with respect to time, the hydrostatic equation, and the first law of thermodynamics. With this estimate of the vertical motion applied to the initial ozonagram sounding, a corrected trajectory value was obtained. The trajectory calculations are justifiable assuming the conservative property of ozone mixing ratio. As linearly interpolated values were not used for trajectory computations, this calculation increased the number of data points between two and three times the original number of ozone data points available for analysis on a pressure surface.

4. Hand Analysis Techniques

The manner in which the data was to be analyzed presented somewhat of a problem. Synoptic models of ozone distribution on a pressure surface have not been published to the author's knowledge--with the exception of the six analyses presented by Berggren and Labitzke (1968). Other investigators have published information about the synoptic distribution of ozone but none have presented distributions on a pressure surface. Pittock (1969) has investigated the synoptic climatology of ozone using statistical methods. His "preliminary results" using 546 Brewer-Mast ozonesonde soundings are primarily in tabular format classed into general synoptic conditions by season. Breiland (1964, 1965, 1967, 1968) studied the distribution of ozone on a sub-synoptic scale primarily using vertical cross sections, time sections, and individual AFCRL network soundings. Sticksel (1966) analyzed the vertical distribution of ozone with data collected at the Tallahassee station of the AFCRL network. Briggs and Roach (1963) and Danielsen (1968) have investigated the ozone distribution, along with other parameters, near the jet stream and the tropopause using aircraft observations.

The first attempt to analyze the plotted data points was inconclusive. Merely constructing isolines of ozone mixing ratio from plotted data points left too much to the imagination. This resulted in inconsistent analyses when compared to the work of the previously mentioned authors and which were also consistent in time.

To overcome this problem the appropriate streamline analysis was put under the plotted ozone data. With the streamlines to guide the analyst in sparse data areas the desired consistency was achieved. To elaborate further, the criteria for placement of isolines of ozone mixing ratio are listed below in order of priority:

a) observed data at map time;

b) data from trajectory calculations as corrected for vertical motion;

c) streamline pattern;

d) vertical motion patterns.

Justification for this priority is obvious in the case of the first two criteria. The choice of the streamline pattern as the third criteria followed by the vertical motion patterns is based on Reed's (1950) conclusion that the horizontal advection contributed 2/3 of the day-to-day change whereas the vertical motions contribute 1/3 of the daily change.

5. Ozone Distribution in the Lower Stratosphere

Before discussing the individual ozone analyses it is worthwhile to compare the twelve-day mean ozone mixing ratio distribution obtained here with distributions prepared by other investigators.

The twelve-day mean ozone mixing ratio computed for the period 29 April to 10 May 1967 is presented in figure 5. It is seen that maximum values at southern latitudes and high altitudes give way to lower values at northern latitudes. The latitudinal gradient reverses between the 30 and 50 mb surfaces with the lower altitudes having higher values at northern latitudes. The latitudinal gradient reverses between the 30 and 50 mb surfaces with the lower altitudes having

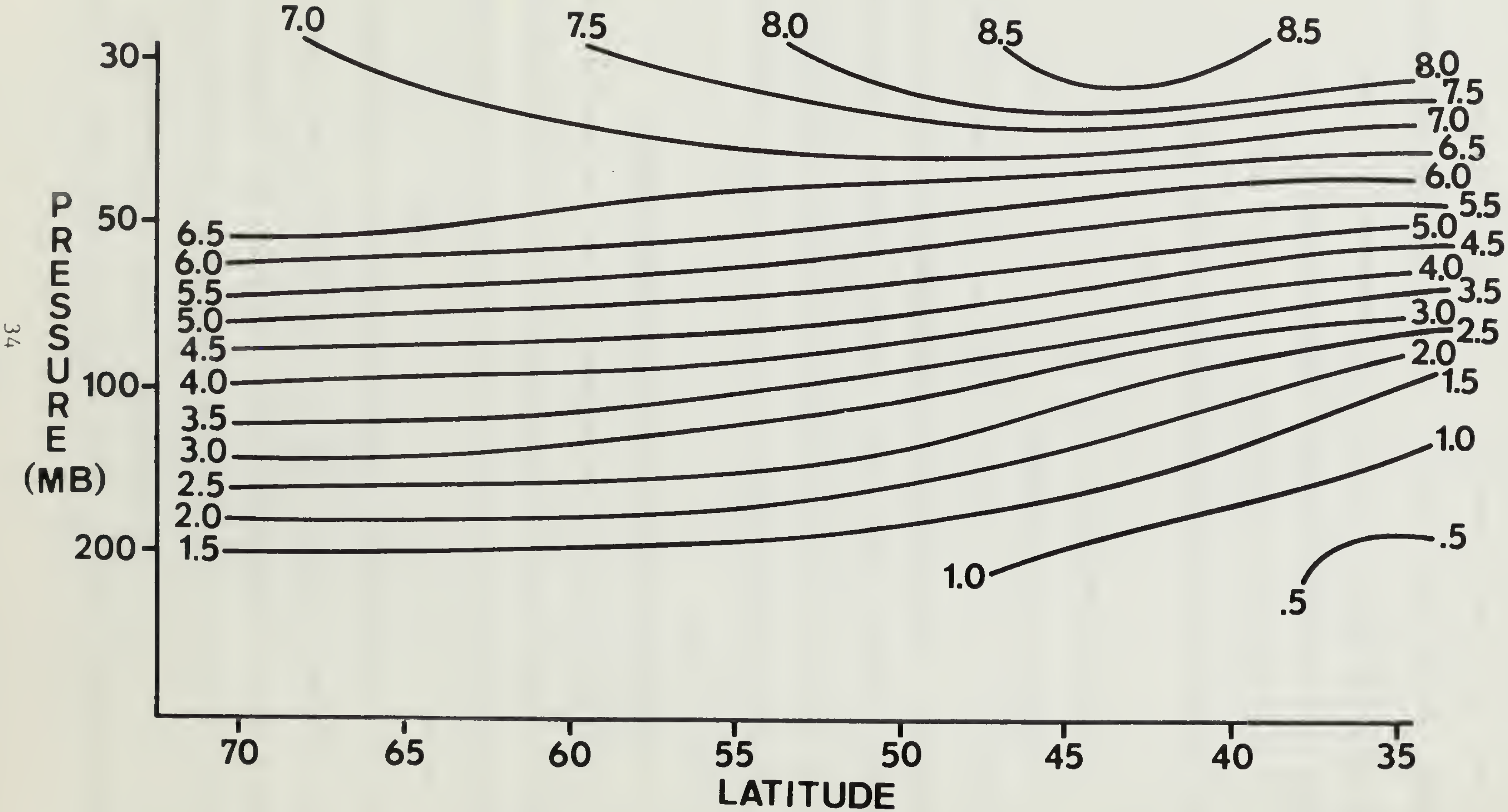

FIG. 5. Twelve-day Zonal Mean Ozone Mixing Ratio (mg g^{-1}) for the period 29 April to 10 May 1963. Computed between longitudes 60W and 140W.

higher values poleward. The slope of the isolines between 50 and 200 mb is markedly downward. In comparison the isolines between 50 and 30 mb are quasi-horizontal between 35N and 50N and rise northward between 50N and 70N.

A direct comparison of ozone mixing ratio values may be made with the values presented by Hering (1966) for the spring of 1963-1964. The two figures are similar except slightly higher values are given here using the 29 April 1963 to 10 May 1963 data. Although Hering uses the same data source as this study, these are the only available, directly comparable, mean ozone mixing ratio values computed from vertical soundings.

Bojkov (1969) presents an annual vertical ozone distribution as determined by the Umkehr method. The units of this distribution are micromillibars and not easily comparable in a quantitative sense. Qualitatively, the same conclusions revealed by the twelve-day mean values are observed in Bojkov's distribution.

Taking into account the information presented by Bojkov and Hering, it is concluded that the twelve-day mean cross section is in agreement with their results and therefore lends credibility to the individual analyses from which the mean distribution was prepared. A representative sample of the distribution of ozone mixing ratio on the 200, 100, 50 and 30 mb pressure surfaces is presented in Appendix A.

The patterns made by the 30 mb ozone mixing ratio isolines are dominated by the effects of the horizontal component of the wind. Ozone-rich air is associated with the northward flow of the subtropical anticyclones forming on the southern edge of a weakening winter circumpolar vortex. Ozone-poor air is associated with the southward flow on the western side of the large scale troughs superimposed on the circumpolar vortex. The two contrasting air masses meet in the middle latitudes and mix. The air, continuing to move eastward, then diverges with relatively ozone-poor air associated with the southward flow on the eastern side of the subtropical anticyclones. Correspondingly, relatively ozone-rich air is associated with the northward flow on the eastern side of the large scale troughs.

If long meridional trajectories form in connection with these cyclonic and anticyclonic wind systems, unusually large values of ozone can be observed at northern latitude stations while lower values are observed several hundred miles east or west of that station. The converse holds true for southern latitude stations. The movement of these wind systems will affect the daily observations at a station in that large day-to-day fluctuations of ozone can be caused by the associated wind shift.

By comparing the vertical motion fields on a pressure surface (not reproduced here) with the ozone mixing ratio distribution, some very interesting observations can be made. The western side of the anticyclones are associated with rising vertical motion, northerly trajectories, and high ozone values while the western side of troughs are associated with sinking motions, southerly trajectories, and low ozone values. Thus it is qualitatively observed that the Reed-Normand effect is operating in the opposite sense than at lower altitudes. The result of this mechanism is in evidence in the mean ozone mixing ratio distribution of figure 5 where the isolines between 30 and 50 mb slope upward at northern latitudes.

Masses of air with similar ozone characteristics can be identified and tracked for periods of several days. Such identifiable air masses seem to be prevalent in the wind fields of the southern anticyclones.

Ozone mixing ratio patterns at 50 mb are similar to the patterns at 30 mb with several exceptions. Streamline patterns at this level reflect the upper, weakening portions of tropospheric systems. Weaker southern anticyclones are also present. The ozone mixing ratio pattern shows ozone-rich air associated with the southward flow that at 30 mb is associated with ozone-poor air. Similarly, ozone-poor air is associated with the northward flow that at 30 mb is associated with ozone-rich air.

A qualitative comparison of the vertical motion patterns with the ozone mixing ratio patterns show the Reed-Normand effect to be operating in the sense originally proposed by Reed (1950) and Normand (1953). The sinking, southward moving air into troughs are areas of high ozone values while rising northward moving air is associated with low ozone values.

Masses of ozone-rich and ozone-poor air can be identified as at 30 mb. However, at both pressure levels it noted that these identifiable ozone masses may be an unnatural consequence of the trajectory supplementation of the data and a weak latitudinal gradient of ozone.

The ozone mixing ratio patterns at 100 and 200 mb are similar to the 50 mb patterns. The effect of the streamline pattern is more pronounced.

The vertical motion pattern on a broad scale reflects the Reed-Normand effect; however, vertical motion maxima with horizontal dimensions on the order 2 or 3 grid intervals are not clearly associated with the Reed-Normand predicted high and low ozone values. This is caused by the density of the ozone data being insufficient to isolate these smaller scale features or by bad ω calculations. Support for this point of view is gained from a calculation of the local change, horizontal advection and vertical advection terms of the Eulerian expansion of the total derivative of ozone mixing ratio with respect to time. This calculation was made on a point by point basis for the entire grid. The results were inconclusive since the error term, computed by assuming ozone is conserved and summing the three remaining terms, was too large in most cases. This indicated a smaller scale ozone distribution that was not able to be identified with the data available.

The ozone analyses prepared for this study compare in a general manner with the distributions used by Breiland (1964, 1965, 1967, 1968), Sticksel (1965), Pittock (1969), Briggs and Roach (1963), Danielsen (1968), and Berggren and Labitzke (1965, 1968) in their respective studies. Only the 200 mb analyses for 3 and 4 May 1963 presented by Berggren and Labitzke (1968) are directly comparable with the analyses compiled here. The analyses are similar in that the major features are the same but minor differences occur with the smaller features. It is significant to note that although the AFCRL data was the initial input in both cases, Berggren and Labitzke used isentropic trajectories to supplement their data points while this study used 24-hour isobaric trajectories corrected for vertical motion.

With the data apparently consistent with the earlier theories, previous investigators, and consistent with respect to other atmospheric parameters it was decided that these analyses would provide an acceptable data base for a quantitative calculation of ozone transport mechanisms.

E. THE OZONE BALANCE EQUATION FOR A VOLUME BOUNDED BY TWO PRESSURE SURFACES

As a first step in evaluating earlier theories of ozone distribution and movement, the calculation of the mean and eddy fluxes of ozone on a synoptic scale are desirable. Considering the minimum amount of ozonesonde observations at a given time, the uncertainty in the synoptic distribution of ozone on a pressure surface, and the uncertainty in the performance of the ozonesonde, an averaging scheme over many data points is preferred as compared to a point by point evaluation. To achieve this, the continuity equation for ozone is written in pressure coordinates as

$$\frac{\partial \chi}{\partial t} = - \mathbb{V}_2 \cdot \nabla \chi - \omega \frac{\partial \chi}{\partial p} + (P - R) + \frac{\partial}{\partial p}\left(K_p \frac{\partial \chi}{\partial p}\right) + \nabla \cdot K_H \nabla \chi. \qquad (3)$$

By putting the advection terms in equation (3) into flux form and taking the area average, equation (3) becomes

$$\frac{\partial \widetilde{\chi}}{\partial t} = - \widetilde{\nabla \cdot \chi \mathbb{V}_2} - \widetilde{\frac{\partial \omega \chi}{\partial p}} + \widetilde{(P - R)} + \widetilde{\frac{\partial}{\partial p}\left(K_p \frac{\partial \chi}{\partial p}\right)} + \widetilde{\nabla \cdot K_H \nabla \chi}. \qquad (4)$$

Now by applying the divergence theorem equation (4) may be rewritten as

$$\frac{\partial \widetilde{\chi}}{\partial t} = \frac{1}{A} \oint \chi \, c_n dl - \widetilde{\frac{\partial \omega \chi}{\partial p}} + \widetilde{(P - R)} + \widetilde{\frac{\partial}{\partial p}\left(K_p \frac{\partial \chi}{\partial p}\right)} - \frac{1}{A} \oint K_H \frac{\partial \chi}{\partial n} dl \qquad (5)$$

where c_n, the component of the wind normal to the boundary, is defined as positive toward the center of the volume.

Along a line let $\chi = \bar{\chi} + \chi'$ where $\bar{\chi}$ is the line average and χ' is the point deviation from the line average. Similarly for averaging operations, equation (5) becomes

$$\begin{aligned}\frac{\partial \tilde{\chi}}{\partial t} = {} & \frac{\bar{\chi}\,\bar{c}_n}{A}\,\Delta l + \frac{1}{A}\,\overline{\chi' c_n'}\,\Delta l - \frac{\partial \widetilde{\omega\chi}}{\partial p} - \frac{\partial \widetilde{\omega^*\chi^*}}{\partial p} \\ & + \widetilde{(P - R)} + \widetilde{\frac{\partial}{\partial p}\left(K_p \frac{\partial \chi}{\partial p}\right)} - \frac{K_H}{A}\,\frac{\partial \bar{\chi}}{\partial n}\,\Delta l,\end{aligned} \tag{6}$$

where it is assumed that K_H is constant on a pressure surface and noted that $\int dl = \Delta l = l_2 - l_1$.

Now average equation (6) between two pressure surfaces, p_u and p_L, to obtain the ozone balance equation

$$\begin{aligned}\overset{\text{(LC)}}{\frac{\partial \tilde{\chi}}{\partial t}} = {} & \overset{\text{(MFLA)}}{\frac{\Delta l}{A}\,\widehat{\bar{\chi}\,\bar{c}_n}} + \overset{\text{(EFLA)}}{\frac{\Delta l}{A}\,\widehat{\overline{\chi'\,c_n'}}} - \overset{\text{(MFLO)}}{\frac{\tilde{\omega}\,\tilde{\chi}}{\Delta p}\Big|_{p_L}} + \overset{\text{(MFT)}}{\frac{\tilde{\omega}\,\tilde{\chi}}{\Delta p}\Big|_{p_u}} \\ & - \overset{\text{(EFLO)}}{\frac{\widetilde{\omega^*\chi^*}}{\Delta p}\Big|_{p_L}} + \overset{\text{(EFT)}}{\frac{\widetilde{\omega^*\chi^*}}{\Delta p}\Big|_{p_u}} + \overset{\text{(PR)}}{\widehat{(P - R)}} + \overset{\text{(SUBLO)}}{\frac{K_p}{\Delta p}\,\frac{\partial \tilde{\chi}}{\partial p}\Big|_{p_L}} \\ & - \overset{\text{(SUBT)}}{\frac{K_p}{\Delta p}\,\frac{\partial \tilde{\chi}}{\partial p}\Big|_{p_u}} - \overset{\text{(SUBLA)}}{\frac{\Delta l}{A}\,\widehat{\left(K_H\,\frac{\partial \bar{\chi}}{\partial n}\right)}},\end{aligned} \tag{7}$$

where the vertical average has been applied to evaluate the terms in a form for numerical computations.

The terms of the equation are:

(LC) the time rate of change of average ozone mixing ratio over the volume;

(MFLA) the mean flux of ozone into the region through the lateral boundaries (the vertical side of the volume);

(EFLA) the eddy flux of ozone through the lateral boundaries;

(MFLO) the mean flux of ozone into the region across the bottom boundary;

(MFT) the mean flux of ozone into the region across the top boundary;

(EFLO) the eddy flux of ozone into the region across the bottom boundary;

(EFT) the eddy flux of ozone into the region across the top boundary;

(PR) the average difference between production and recombination of ozone for the whole volume;

(SUBLO) the flux of ozone into the region, across the lower boundary, due to sub-grid scale diffusion;

(SUBT) the flux of ozone into the region, across the upper boundary, due to sub-grid scale diffusion;

(SUBLA) the flux of ozone into the region through the sides due to sub-grid scale diffusion.

All the terms have units of micrograms per gram per day. This unit is convenient in view of the time scale of the observations. In addition, the value of the ozone mixing ratio at a given height appears as a variable on the ozonagram.

Computer programs were written to evaluate all terms on a daily basis for the 30/50, 50/100, and 100/200 mb volumes with the exception of the production and recombination term and the sub-grid scale diffusion terms.

Brewer and Wilson (1968) have evaluated the meridional net ozone production rate for the winter and summer season from the equator to sixty degrees latitude and between 200 and 30 mb. Since the evaluation of the PR term would require a complete and separate treatment, Brewer and Wilson's results are accepted for use in this calculation. To compute an estimated value for the net ozone production rate, a rough interpolation between seasons and pressure surfaces followed by an estimated latitudinal average was necessary. The appropriate unit conversions were made. These values will be presented with the results of the ozone balance equation in a later section.

The sub-grid scale diffusion terms are expected to be small when compared to the magnitude of the other terms. Lacking a reliable estimate of the eddy-diffusion coefficients of ozone mixing ratio, these terms were not calculated.

The initial attempt to evaluate the ozone balance equation by using the observed wind to directly evaluate each term resulted in unusually large errors that could be traced to the fluctuations of the mean terms. To improve upon these results the mass continuity equation was solved for the mean normal component of the wind at the side boundaries and in terms of the computed $\tilde{\omega}$ fields. The evaluation of the mean normal component of the wind can be obtained by applying the divergence theorem to the area-averaged continuity equation and solving the following form,

$$\widetilde{\overline{c}}_n = \frac{A\,(\tilde{\omega}_{p_L} - \tilde{\omega}_{p_u})}{\Delta l\,(p_L - p_u)} \quad . \tag{8}$$

Although this scheme assured the conservation of mass it increases the dependence of the calculations on the computed vertical motions as compared to directly observed winds.

III. EVALUATION OF THE TERMS OF THE OZONE BALANCE EQUATION

Without further modification the terms of the ozone balance equation were computed. The daily calculations tended to fluctuate rapidly and may be unrealistic. Considering the magnitude of the error term in comparison with the magnitude of the other terms, it was decided to present the ten-day mean values of the terms. These values are given in tabular format in Table II. In the computed format, the individual mean and eddy terms cannot be compared directly. To effect a comparison the mean terms must be summed thereby giving the net effect of the mean terms on the volume.

To obtain a form in which the mean horizontal and vertical flux may be directly compared to the eddy flux terms, the sum of the mean terms, as presented in Table II, may be computed in another manner. The area-averaged continuity equation can be written as a representation of the net effect of the mean terms at the middle pressure between two pressure surfaces. By applying a vector identity and the divergence theorem, the first and third terms on the right hand side of equation (6) can be written as

$$\mathrm{SUM} = \overline{c_n}\,\overline{\chi}\,\frac{\Delta l}{A} - \tilde{\omega}\,\frac{\partial \tilde{\chi}}{\partial p} - \frac{\tilde{\chi}}{A}\oint c_n\,\delta l. \tag{9}$$

By combining like terms and putting into computational format, equation (9) becomes

$$\mathrm{SUM} = \overset{\text{(MLAD)}}{(\overline{\chi} - \tilde{\chi})\,\frac{\Delta l}{A}\,\overline{c_n}} - \overset{\text{(MVAD)}}{\tilde{\omega}\,\frac{\partial \tilde{\chi}}{\partial p}}. \tag{10}$$

Key: See Section II-E

SUM = MFLA + MFLO + MFT

AS COMPUTED:

Volume	(LC)		(MFLA)	(EFLA)	(MFLO)	(MFT)	(EFLO)	(EFT)	(PR)	(ERROR)
30/50 mb	-173	=	+ 184	+ 54	-688	+696	- 67	-50	-125	+177
50/100 mb	-517	=	+1102	- 46	-921	+275	-103	+27	- 44	+806
100/200 mb	-273	=	- 229	-321	- 47	+461	-155	+51	- 16	+ 17

COMPARISON FORMAT:

Volume	(LC)		(SUM)	(EFLA)	(EFLO)	(EFT)	(PR)	(ERROR)
30/50 mb	-173	=	+192	+ 54	- 67	-50	-125	+177
50/100 mb	-517	=	+456	- 46	-103	+27	- 44	+806
100/200 mb	-273	=	+185	-321	-155	+51	- 16	+ 17

TABLE II. The Ten-day Mean Ozone Balance for Indicated Volumes. Symbols for each term given in Section II-E. Units are micrograms per gram per day x 10^4.

The two terms are defined as:

(MLAD) the mean advection through the lateral boundary;

(MVAD) the advection into the volume due to the mean vertical motion.

The two terms of equation (10) were evaluated using the same input values as for the evaluation of equation (7). The results of the evaluation of equation (10) are presented in pictorial form in figure 6 with the local change and eddy terms as evaluated by equation (7). The net production term is the same in both cases. The error terms in each evaluation are different only to a small degree. It is therefore concluded that the computational scheme used to arrive at the value of the mean terms is a reasonable evaluation of the input parameters.

With the mean and eddy ozone flux terms in a format for comparison and some degree of confidence in the method of evaluation of the mean terms, it is appropriate to discuss the individual terms of the ozone balance as presented in figure 6.

A. THE LOCAL CHANGE TERM

Computation of this term was made directly from the distribution of ozone on a pressure surface and therefore is not affected by other computed parameters. A rough check on the value of this term was made by comparing the computed result, multiplied by 100, with a 100-day difference between area averages estimated from Godson's (1960) mean distribution of total ozone. This period falls after the March maximum of total ozone shown by Godson and the sign of the mean local change term is negative. With these observations in mind, the 100-day period between May and August was chosen for comparison.

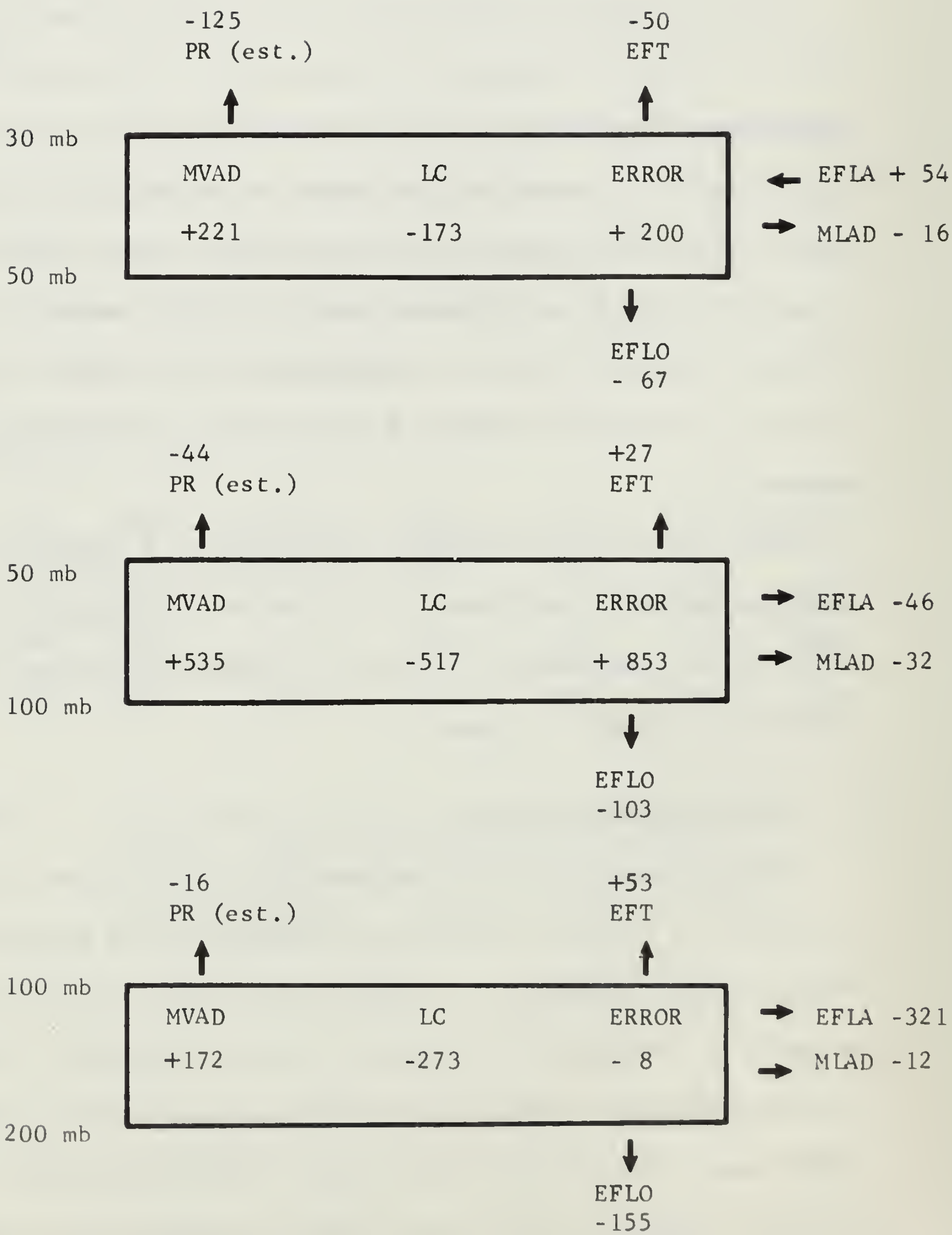

FIG. 6. Ten-day mean ozone balance in advective format. Symbols for each term given in Section II-E and Section III. Units are micrograms per gram per day x 10^4.

After making the appropriate unit conversions, the mean local change term accounted for 85 percent of the change in Godson's distribution. Thus the rate of decrease of ozone within the volume for this period appears representative of the seasonal trend.

It is significant to note that the maximum of total ozone in the atmosphere is observed in March and a net hemispheric decrease is observed after that month (Godson, 1960). The dynamic mechanisms or conditions that created the maximum conditions, particularly at the northern latitudes, must have ceased operating. One may speculate that the mechanism to accomplish this winter-spring northern maximum is the long meridional trajectories set up by the breakdown of the polar night vortex associated with the mid-winter "sudden warmings". Further, the dynamic meachnisms or conditions now operating must be serving to deplete the atmosphere of ozone.

B. THE MEAN TERMS

The mean advection through the lateral boundaries can only be evaluated as to direction into or out of the volume. In view of this limitation, the only point to be made is a mention that the term in each volume is contributing a small net loss to the volume and the mean terms (MLAD) are smaller than their respective eddy terms (EFLA) (see fig. 6).

On the other hand, the mean advection in the vertical shows a consistent downward motion through the three volumes of interest. In the two upper volumes the mean vertical advection is the dominant term.

C. THE EDDY TERMS

In the 30/50 mb volume the lateral eddy flux term (EFLA) is an input to the volume and agrees with the earlier qualitative observation that the anticyclones on the southern edge of the circumpolar vortex appeared to be transferring ozone northward. The magnitude of the lateral eddy flux term (EFLA) is small relative to the observed change within the volume.

The lateral eddy flux terms at 50/100 and 100/200 mb serve to deplete the volumes of ozone. The value of the term at 50/100 mb is small relative to the mean vertical advection and the vertical eddy flux across the 100 mb surface. In the 100/200 mb volume the lateral eddy flux term is the largest term in the ozone balance. An interesting and unexplained question is the ultimate destination of the ozone advected out of the volume of computations by the lateral eddy flux.

The vertical eddy flux terms show a consistent downward motion from 50 to 200 mb and are a measure of the vertical motion portion of the Reed-Normand effect. At these levels the direction is consistent with that proposed by Reed and Normand. At the 30 mb surface the direction of the vertical eddy flux is upwards. This can be traced to the northward, rising ozone-rich air associated with the western side of the anticyclones at this level. This is consistent with the earlier section in which the Reed-Normand effect was observed to be operating in the opposite sense as observed in lower altitudes.

By comparing the vertical eddy flux at the upper and lower surfaces at each volume, it can be seen that these eddies contribute a net loss of ozone to each volume. Another interesting, unanswered question is posed by this observation. If a given amount of ozone is going downward into a trough, what physical process or dynamic mechanisms are operating to prevent an equal amount of ozone from proceeding back upwards to the ridge? The answer probably lies with a smaller scale turbulence phenomenon not yet identified much as the energy cascade in the lowest level of the atmosphere.

The vertical eddy flux terms are the most significant of all the terms computed in the ozone balance. This conclusion is attributed to the fact that the vertical eddy flux computed for this particular area's pressure surfaces may be representative of the whole global pressure surface in question. This cannot be said of the lateral eddy flux terms. As is to be described in a later section, the magnitude of the mean terms is subject to question.

D. THE PRODUCTION AND RECOMBINATION TERM

In comparison with the magnitudes of the other terms, the production and recombination term as estimated from Brewer and Wilson's (1968) values does not have a significant effect except in the 30/50 mb volume. Here the magnitude is about 2/3 of the local rate of change. In future investigations this must be taken into consideration.

E. THE ERROR TERM

The error term in Table II and figure 6 is the value that must be added to the local change in order to equal the sum of the mean, eddy and net production terms. In numerical computations of this sort, errors are expected because of data uncertainties and since many approximations are made to facilitate the evaluations of the individual terms and their components.

The mean errors in each volume are of the order of magnitude of the largest term. The mean errors at 30/50 and 50/100 mb are representative of the daily errors at these levels from which the mean error was derived. This is not true at the 100/200 mb volume where the daily errors average one order of magnitude larger than the ten-day mean error. Thus it is noted that the ten-day mean error is unrepresentative of the error in the 100/200 mb volume and, further, that the calculations here suffer from the same problems as the upper two volumes.

Considering that the error in all three volumes indicates too much ozone on the right hand side of the balance, the failure to compute the sub-grid scale diffusion terms has to be considered. It is expected that these terms will evacuate ozone from the volume with a magnitude considerably smaller than the magnitude of the error. Therefore it is improbable that the failure to calculate these terms contributes much to the error.

The first serious possible source of error is that the ozone mixing ratio distributions on the pressure surfaces are not accurate enough for the calculations. Only by having a more dense data network will this ever be known for sure. The probability that this is the major source of error is diminished considering the consistency in daily analyses and the consistency of the distributions with the theories and distributions advanced by earlier investigators.

The second and most probable source of error is in the evaluation of the vertical motions. As was noted earlier the evaluation of the ozone balance equation is heavily dependent upon vertical motions. While a consistent error in the vertical motions will not affect the eddy terms greatly, it will have considerable effect on the mean terms.

Since the continuity equation was used to evaluate the mean flux through the lateral boundaries, this term is affected also. This line of reasoning is consistent with the fact that the mean terms in most cases are large compared to the rest of the terms as can be seen in Table II and figure 6. It can be reasoned that a consistent error in the vertical motion at a particular pressure level could affect each of the mean terms enough to produce a significant error.

The size of the error in the vertical motion does not have to be very large to seriously affect the calculations. To see this, consider the vertical motion scheme used in the computations and presented as equation (1). The diabatic heating term is the term most subject to question as its values determined by any method are highly uncertain. This is attributed to the lack of an accurate method to check the results of the determination of the diabatic heating. If the area average of equation (1) is taken and it is assumed that the correction to the vertical motion ($\tilde{\omega}_{corr}$) is a function only of the correction to the diabatic heating term (corr) then it can be written that approximately,

$$\tilde{\omega}_{corr} = \frac{-corr}{\alpha/c_p}. \tag{11}$$

A representative value of area averaged diabatic heating at 75 mb as estimated from Kennedy's (1965) results can be considered -.35 degrees per day. If this were incorrect by an arbitrarily selected $\pm$ .2 degrees per day, then the evaluation of equation (11) using representative values for evaluation of the specific volume would produce

$\tilde{\omega}_{corr} = \pm$ .24 mb per day, or about a 25 percent error in the mean vertical motions.

The mean vertical advection of ozone across a pressure surface is represented by $\tilde{\omega} \frac{\partial \tilde{\chi}}{\partial p}$ in equation (10). Evaluating the effect of $\tilde{\omega}_{corr}$ on this term using representative values of $\tilde{\chi}$ produces uncertainty in this term of approximately 210 x 10^{-4} micrograms per gram per day. It is easily seen from equations (7) and (8) how this would affect the mean flux through the lateral boundaries as presented in Table II. The value of the mean advection through the lateral boundary is small (fig. 6). Therefore, it is reasoned that in this form the mean lateral advection is not a major source of error. Similar arguments can be made for the 30/50 and 100/200 mb mean terms.

It can be concluded that the uncertainty in the evaluation of mean stratospheric vertical motions is large enough to result in the observed magnitude of the error to the ozone balance equation through evaluation of the mean terms. Implied from the excess of ozone represented by the error term is that the net input to the particular volume by the mean terms, particularly the mean vertical flux (or advection) terms, is too large although the sense of the individual mean terms is probably correct.

IV. ADDITIONAL CALCULATIONS

An attempt was made to support the results of the qualitative evaluation of the distributions of ozone on a pressure surface and the terms of the ozone balance by calculating the correlation coefficients of the meridional wind and ozone mixing ratio and the correlation coefficients of the vertical motion and the ozone mixing ratio. The calculations are presented as figures 7 and 8. For the number of data points involved a correlation coefficient greater than .2 is statistically significant at the 95 percent level of confidence.

The correlation coefficient for two variables is defined as the covariance divided by the variance. It follows then that the sign of the correlation coefficient is also the sign of the covariance. This relationship allows comparison with the eddy terms of the ozone balance.

Looking at figure 7, it can be seen that there are non-significant values of the correlation coefficients for most of the 30 and 50 mb surfaces during the ten-day period. This is consistent with the small values of the lateral transport computed by the ozone balance. The ozone distributions observed on the pressure surfaces are evaluated as having been spread out by the weak wind fields at these levels and accomplishing little transport. The positive area at 30 mb and 70N latitude is not conclusive since this area was determined by one data point. It is consistent, however, with the assumption that ozone-poor air is advected southward by the horizontal eddy flow at this altitude.

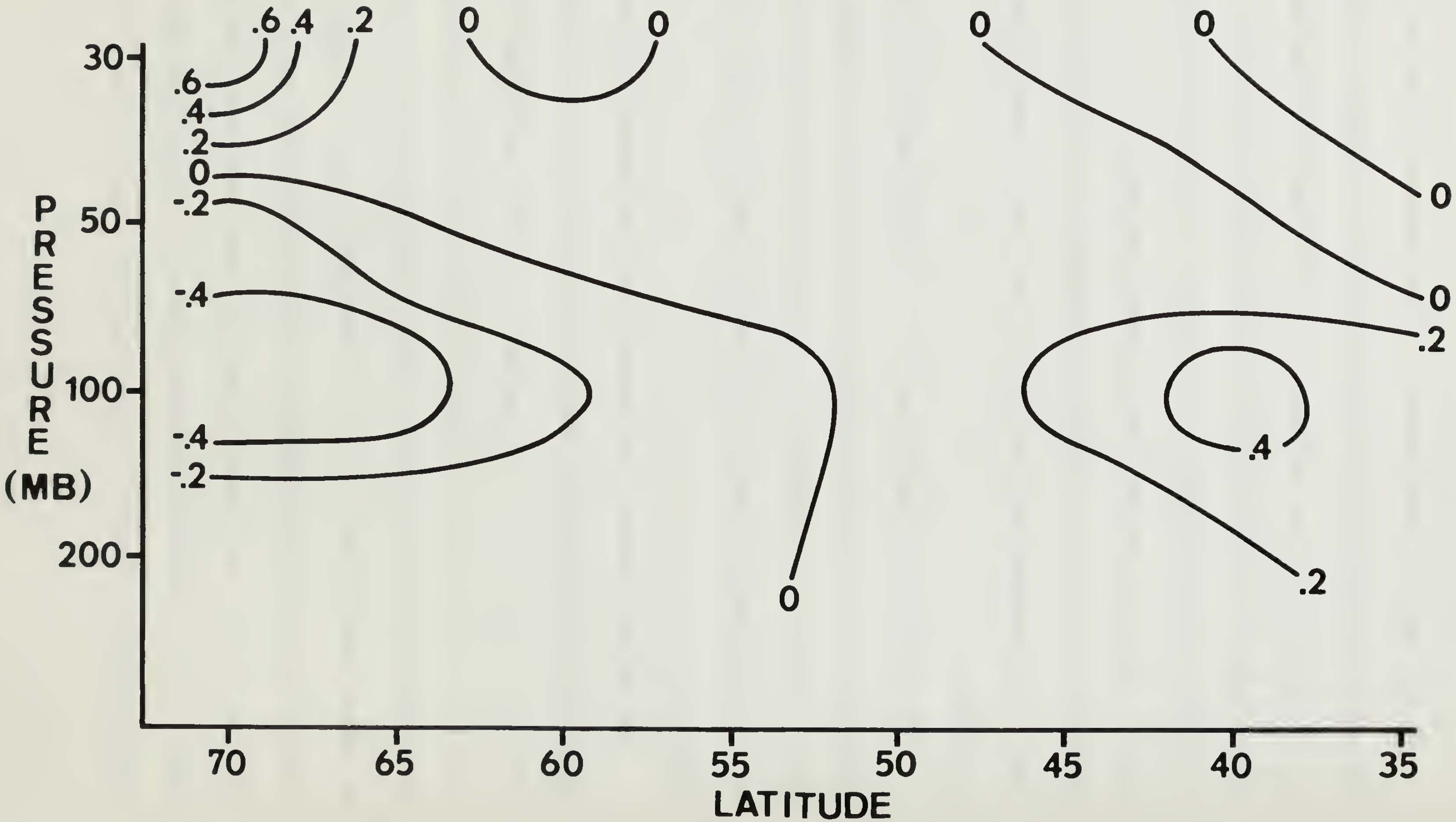

FIG. 7. Ten-day Mean Correlation Coefficients Between Meridional Wind and Ozone Mixing Ratio for the Period 30 April to 9 May 1963. Computed between longitude 60W and 140W.

At the 100 mb level the negative values to the north are associated with high ozone values being moved southward by the horizontal eddy flow. The positive values to the south are an indication of a counter-gradient transport (see fig. 5) of high ozone values northward and low ozone values southward by the horizontal eddy flow. To evaluate the cause of the counter-gradient transport, the vertical motion patterns (not presented here) were compared with the streamline patterns. It was observed that in many cases the maximum sinking areas connected with ozone-rich air were slightly (45 degrees in phase angle) to the east of the base of the trough. This offset of the vertical motion pattern from the trough line has been discussed by Ohring and Muench (1960). The sinking consequently offset the entire ozone mixing ratio pattern to the east and set the conditions for counter-gradient flow by the horizontal eddies.

The results at 200 mb were expected to be more conclusive than shown in figure 7. The values reflect the stronger relationships at 100 mb, however, they are not at the 95 percent level of significance. Without further calculations, a definite conclusion cannot be advanced.

The results of figure 8 strongly support earlier contentions. The weak positive values are associated with upward eddy transport of high ozone values by the anticyclonic activity at 30 and 50 mbs. The high negative values at the southern latitudes indicate the expected downward transport of high ozone values. These latitudes coincide with the base of the troughs on the circumpolar vortex and hence general areas of maximum sinking motions. At 100 mb the secondary maximum of significant negative values at the northern latitudes are evaluated as downward transport of high ozone values by the weaker semi-permanent

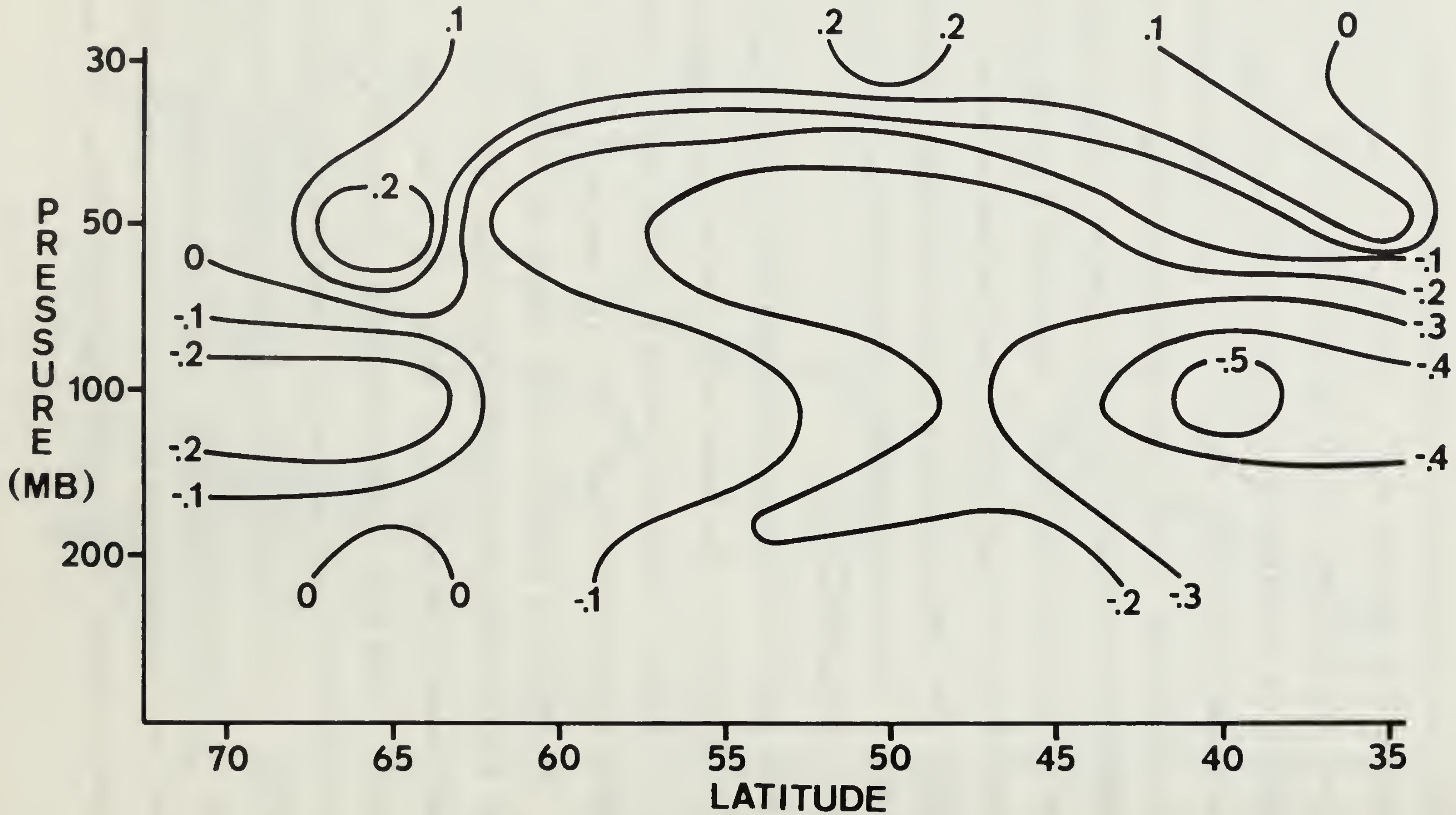

FIG. 8. Ten-day Mean Correlation Coefficients Between Vertical Motion and Ozone Mixing Ratio for the Period 30 April to 9 May 1963. Computed between longitude 60W and 140W.

trough of the circumpolar vortex. Based on the values at 200 mb it is further reasoned that the vertical eddy flux of ozone across the tropopause is taking place in connection with the maximum sinking areas near the base of the transient troughs on the circumpolar vortex.

In the recent past several investigators (Newell 1961, 1962, 1963, 1964; Mahlman, 1966, 1967; and Molla and Loisel, 1962) have used the latitudinal covariance of the meridional and vertical wind components $\overline{(v'w')}$ as an indication of stratospheric eddy transport processes. As a further check on the validity of the terms of the ozone balance equation, it was decided to compare $\overline{(v'w')}$, by using correlation coefficients, with the information prepared for this study. This distribution is presented in figure 9.

The negative values of the correlation coefficients are associated with a northward and downward transport while the positive areas indicate a southward and downward transport. The information from fig. 9 is in agreement with the sense of the vertical eddy flux terms of the ozone balance, and the ozone distribution on a pressure surface except at the 30 mb surface. At this level the mean ozone mixing ratio, streamline and vertical motion patterns show a northward and upward flow of high ozone values. This was confirmed by a vertical eddy flux out of the 30/50 mb volume across the 30 mb surface and figure 8. The vertical eddy flux is a direct calculation of transport as compared to the correlation coefficients which are only an indication of transport. This implies that the correlation coefficient must be on the order of magnitude of .5 or greater to give confidence in the conclusion that this term is in fact an indicator of transport.

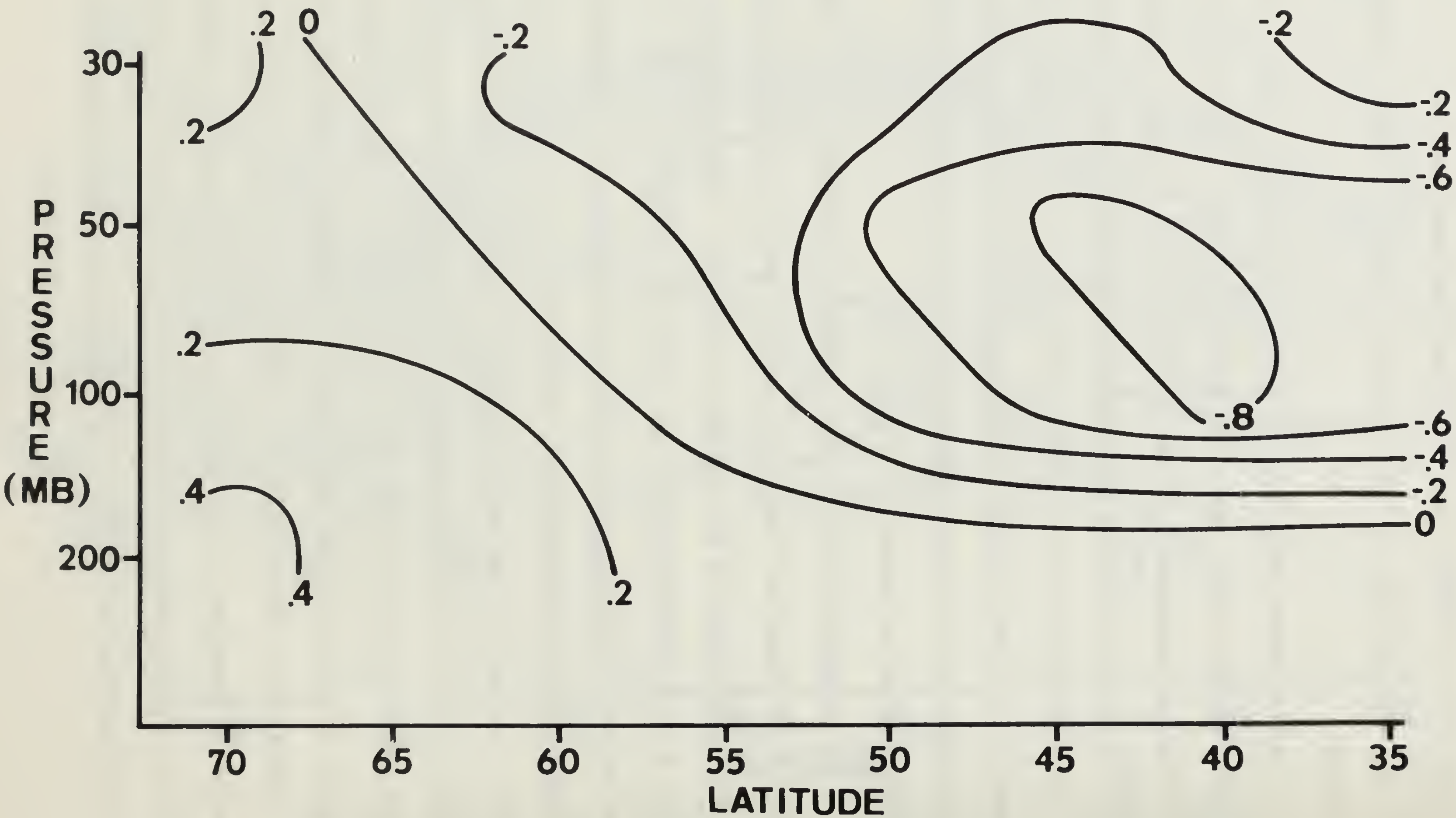

FIG. 9. Ten-day Mean Correlation Coefficients Between Meridional Wind and Vertical Motion for the Period 30 April to 9 May 1963. Computed between longitude 60W and 140W.

V. SUMMARY AND CONCLUSIONS

Even though the magnitude of the mean terms is subject to question, the distribution of ozone on a pressure surface, the ozone balance, and the supporting calculations allow several general conclusions to be drawn.

1. It is possible to construct the distribution of ozone on a pressure surface from the Regener ozonesonde data and then perform numerical calculations that provide reasonable results.

2. During this time period, the atmosphere is past its yearly maximum content of ozone at northern latitudes. There is no evidence to support a large northward movement of ozone at this time.

3. During the ten-day period in question, ozone content of the volume is decreasing. To accomplish this, the mean vertical motions, supported by the vertical eddy transport at 50, 100, and 200 mb, are moving ozone downward to the tropopause level where the vertical eddy transport associated with the transient eddies on the circumpolar vortex then move the ozone into the troposphere for eventual destruction.

4. The vertical eddy transport is a measure of the vertical part of the Reed-Normand effect. This mechanism is operating in the sense as proposed between 50 and 200 mb. At 30 mb the Reed-Normand effect is operating in the opposite sense with ozone-rich air being advected upward.

APPENDIX A

STREAMLINE AND OZONE MIXING RATIO ANALYSIS FOR 3 MAY 1967

This appendix presents a representative sample of the distributions of ozone mixing ratio on a pressure surface in the lower stratosphere. To understand the effect of the wind field upon the ozone distribution, the appropriate streamline pattern is presented.

The choice of date was made to facilitate comparison with the patterns published by Berggren and Labitzke (1968).

FIG. A-1. The 200 mb Streamline Analysis for 3 May 1963.

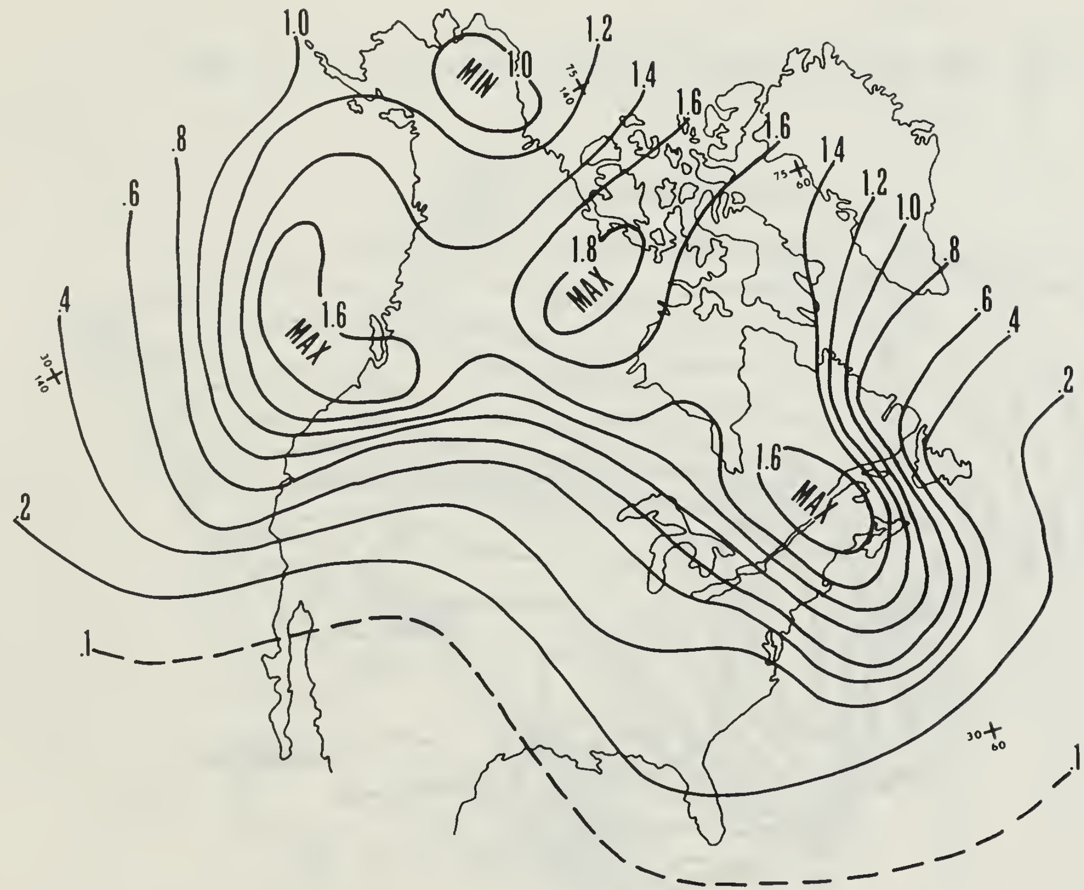

FIG. A-2. The 200 mb Ozone Mixing Ratio (mg g^{-1}) Analysis for 3 May 1963.

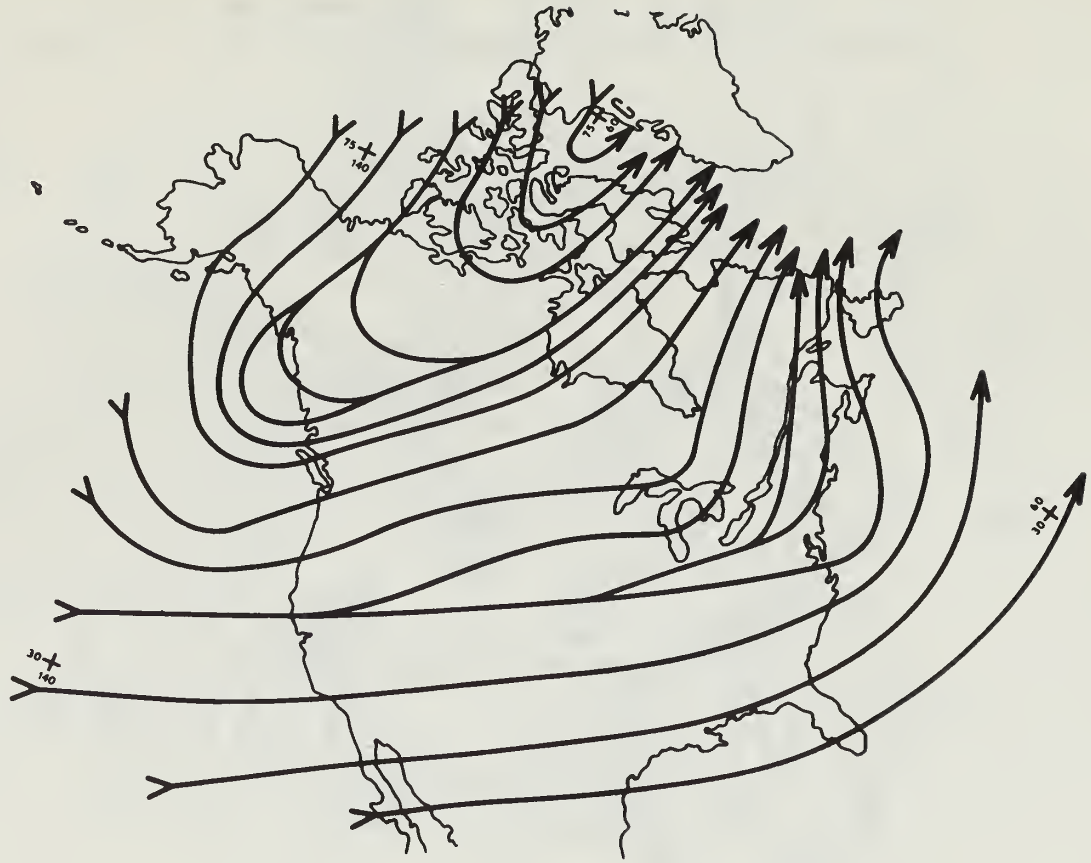

FIG. A-3. The 100 mb Streamline Analysis for 3 May 1963.

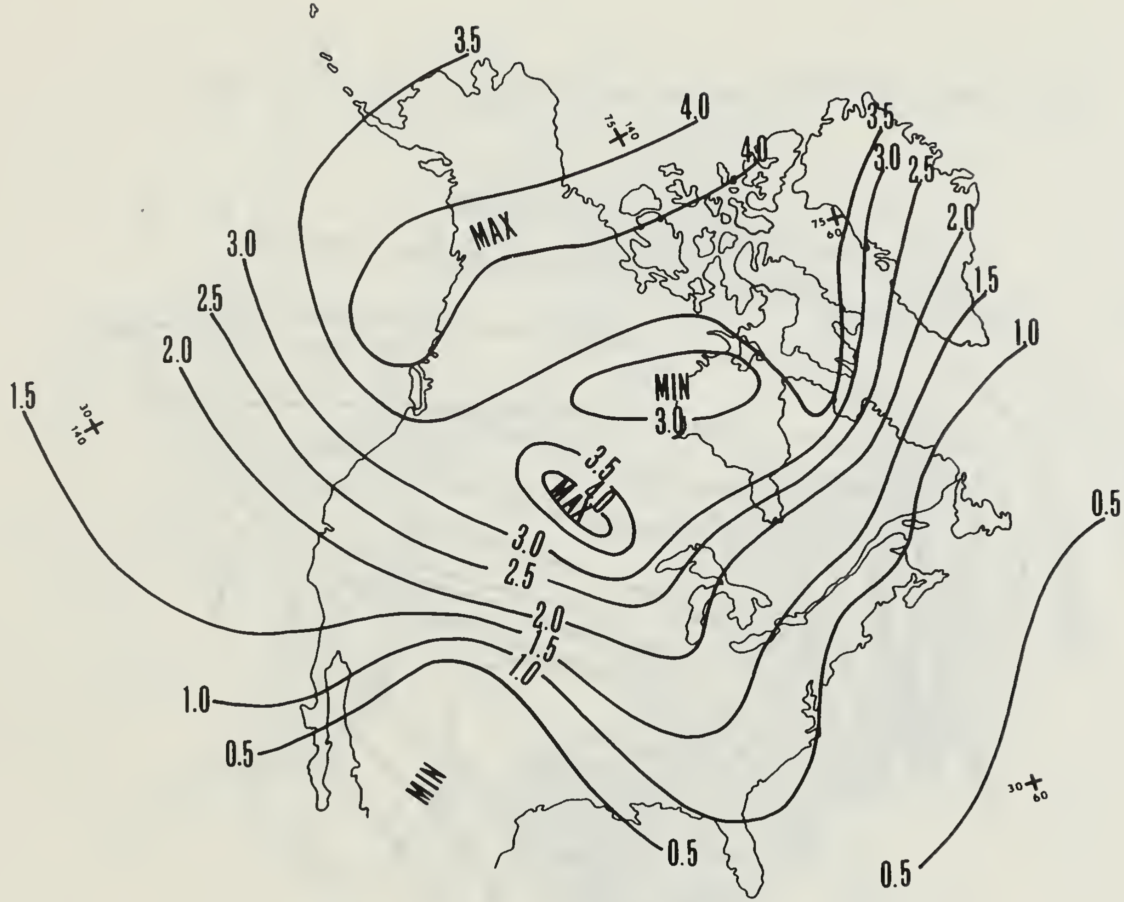

FIG. A-4. The 100 mb Ozone Mixing Ratio Analysis (mg g^{-1}) for 3 May 1963.

FIG. A-5. The 50 mb Streamline Analysis for 3 May 1963.

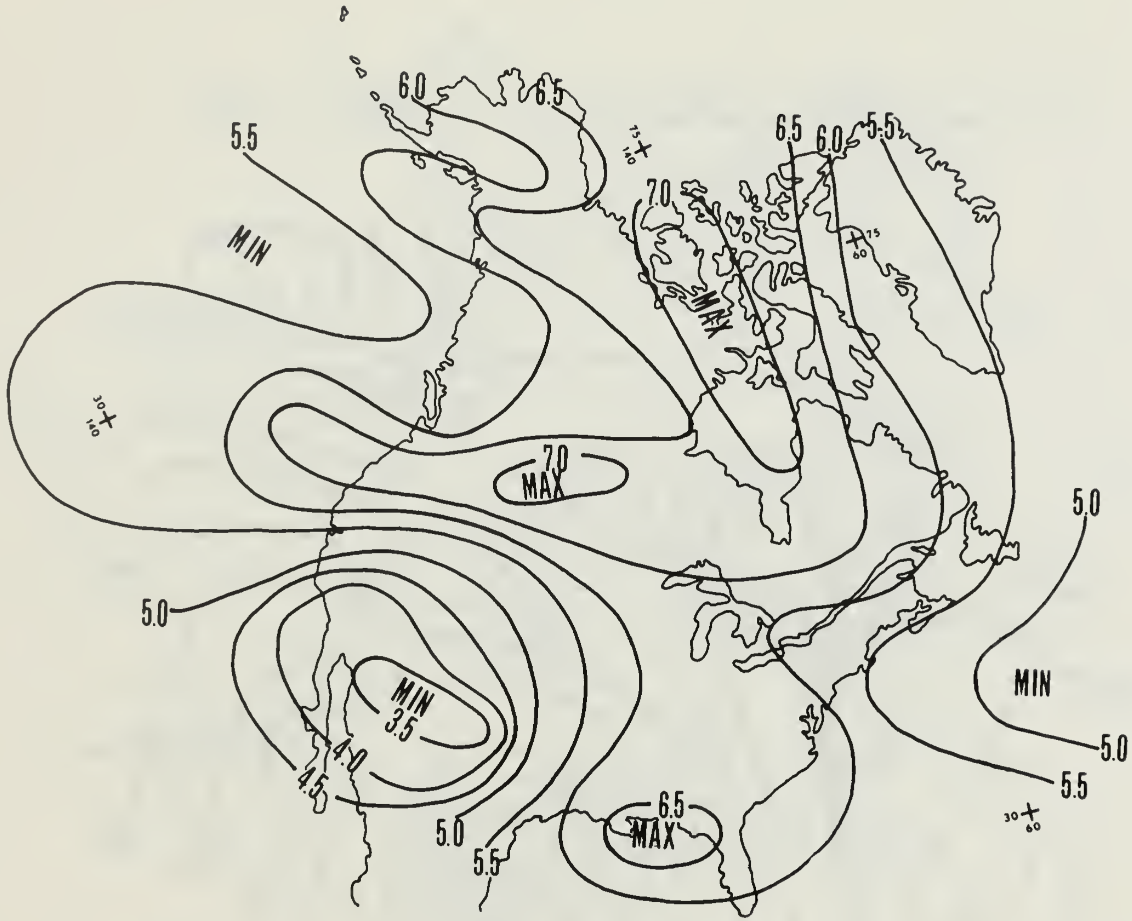

FIG. A-6. The 50 mb Ozone Mixing Ratio (mg g^{-1}) Analysis for 3 May 1963.

FIG. A-7. The 30 mb Streamline Analysis for 3 May 1963.

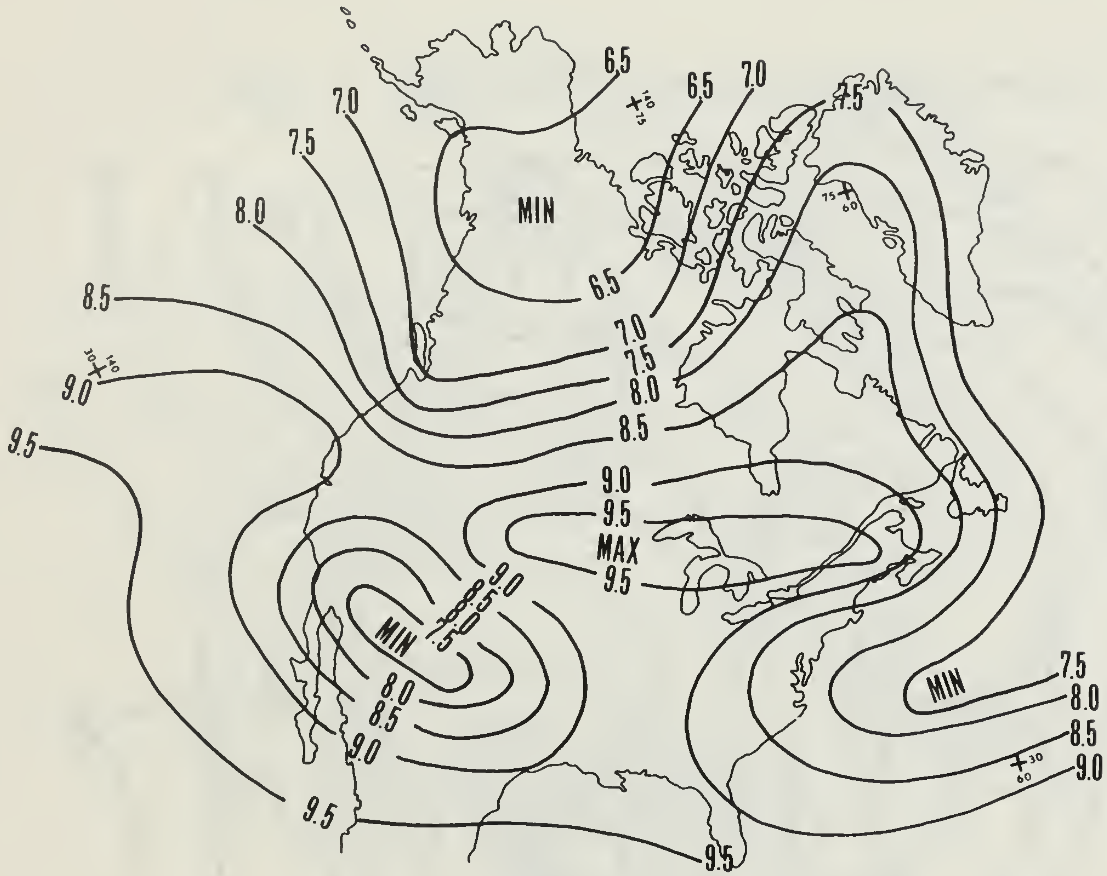

FIG. A-8. The 30 mb Ozone Mixing Ratio (mg g^{-1}) Analysis for 3 May 1963.

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13. ABSTRACT

The vertical distribution of ozone was observed by the Air Force Cambridge Research Laboratories North American ozonesonde network from 29 April 1963 to 10 May 1963 on a daily basis. This observed data, supplemented by twenty-four hour trajectory calculations, was used to prepare distributions of ozone mixing ratio on the 200, 100, 50, and 30 mb surfaces. The ozone distributions were evaluated for the ozone transport by mean and eddy motions in the lower stratosphere. The results of these calculations show the magnitude of the horizontal and vertical transport of ozone into and out of the 30/50, 50/100, and 100/200 mb volume over North America. The mean vertical transport, supported by the vertical eddy transport, is combined to move the ozone primarily downward through the stratosphere. At the tropopause the vertical eddies transport the ozone to the troposphere for eventual destruction.

14 KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
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Ozone						
Atmospheric Circulation						
Advection						

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